

PROSPECTS FOR RF APPLICATION OF HIGH TC SUPERCONDUCTORS

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Abstract

RF measurements from 0.1 to 150 GHz on high Tc superconductors in bulk ceramic and thin film forms show that losses at the present level of materials preparation techniques are high and increase rapidly with increasing RF surface magnetic field. At this stage the new superconductors are not competitive with Nb at liquid He temperature or with copper at room temperature. On the other hand, superior RF properties revealed by measurements on crystal YBaCuO indicate that the problems are not intrinsic to high Tc superconductors. We are therefore encouraged to expect that as new fabrication techniques emerge to conquer the prevalent problems of grain orientation, intergrain impurities, impurity phases, and oxygen stoichiometry, microwave applications of high-Tc oxide superconductors will eventually be realized.

Introduction

Prospects for eventual application of high Tc superconductors (HTS) depend on several factors. Most obvious are the properties we know very well to be crucial to the good performance of RF cavities: the minimum achievable surface resistance, the response of the resistance to application of the RF fields, the maximum possible surface magnetic and electric fields. As we shall present in depth, at this stage of materials development, these properties are substantially inferior to that of Nb, the current favorite for RF superconductivity applications. However, this situation could be predominantly influenced by processing parameters, so that it may be too early to make a fair assessment of RF application potential. We must therefore strive to establish the intrinsic properties and attempt to assess whether they offer any potential advantages which are now in doubt because of materials uncertainties. We must also consider to what extent the fundamental properties which are special to the new superconductors could ultimately be responsible for the poor RF properties. Finally, application prospects must be judged from an estimate of the benefits from increased operating temperature and possible increased operating fields that could be realised if HTS could eventually become available with engineering properties equal to or superior to existing materials. Could the leverage of HTS in future large scale applications be sufficient to displace Nb?

Survey of RF experiments

A tremendous experimental effort is underway to study the RF properties of HTS.[1-11]. Laboratories involved are Argonne, Cornell, David Sarnoff Research Center, Los Alamos, MIT, Naval Research Laboratory, Northeastern, Sandia/Wisconsin, Stanford/UCLA and Wuppertal. RF properties interesting to accelerator applications are best studied using a resonant cavity in the TM010 mode, the entire surface of which is made from superconducting material. However, at the present stage, materials techniques are not sufficiently mature to allow this; instead samples of HTS in forms most suitable to the particular preparation technique are inserted into a host cavity or micro-strip line resonator out of normal metals such Copper, plated gold or superconductors such as Nb or Pb-on-Cu.

RF results on HTS

Most information on RF properties is available on YBCO. An overview of low field surface resistance results on bulk, thin film and crystal YBCO is presented in Fig. 1 at 77 K and in Fig. 2 at 4.2 K [1-11] and compared to the resistance of room temperature copper as well as to the 4 K resistance of Nb. The bottom of the vertical scale is characteristic of residual losses in Nb ($10^{-8} \Omega$), which are essentially frequency independent. Early results on Bi- and Tl- compounds are summarised in Table 1.

Like for the ordinary superconductors, there is an underlying frequency dependence to the 77 K surface resistance, which at this stage may be consistent with f^2 . What is surprising is that there is also an f^2 -like frequency dependence for the residual losses at 4 K, which is not observed in conventional superconductivity.

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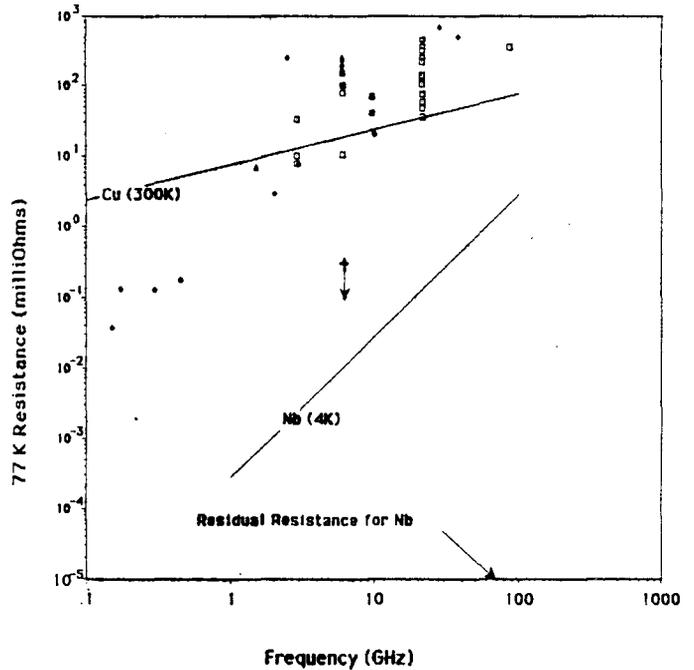


Fig. 1. Survey of 77 K RF surface resistance measurements from 0.1 to 150 GHz, compared with 4K Nb and room temperature copper. The arrow on the 6 GHz crystal data shows that the point is an upper bound.

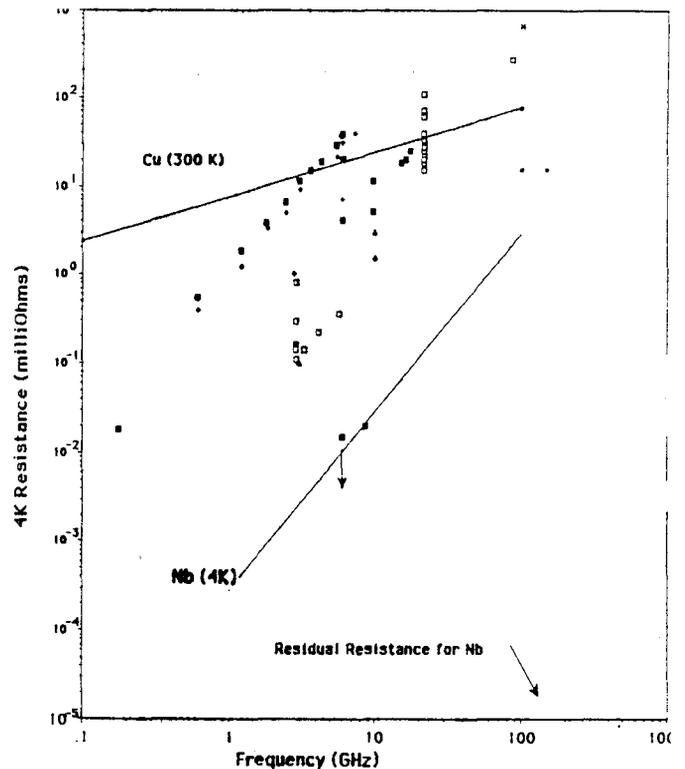


Fig. 2. Survey of 4 K RF surface resistance measurements from 0.1 to 100 GHz, compared with 4K Nb and room temperature copper.

Table 1 :RF Resistance of Bi- and Tl- Compounds

Lab	Frequency (GHz)	T (K)	RF Resistance ($10^{-3}\Omega$)	Compound
Argonne	2.65	77	70	Bi- (80K)
"	3.65	77	70	Bi- (80K)
"	2.65	4.2	7	"
"	3.65	4.2	9	"
Argonne/UC-San Diego	29	4.2	45	oriented Bi-Pb-doped
Los Alamos	3.	4.2	4.4	Tl-
NRL	18	77	30	Tl- (120 K)
"	18	20	10	"
UCLA/Stanford	102	4.2	800	Bi-Crystal ab plane
Wuppertal	22	20	60	Bi- (80K)

At a fixed frequency, the wide scatter in both figures clearly reflects the prevailing influence of process parameters. At 77 K, the resistance values closest to those for Nb (4K) are with crystals, and again, at 4.2 K, the values closest to Nb (4K) are with crystals or with epitaxial thin films on SrTiO₃. Thus the best measured resistance at liquid nitrogen temperature is a factor of 30 higher than for Nb at 4 K and a factor of 70 lower than room temperature copper. Keeping in mind a factor of 25 gain in Carnot efficiency between 4 K and 77 K, these results are very encouraging. Arrows on the crystal data indicate that values obtained so far are only limits set by the prevailing measurement sensitivity, which in the case of crystals is a difficult problem because only mm² size samples are available.

The shape of the RF transition is another important indicator to consider, and is strongly influenced by process technique. For a long time, all measurements, including the first crystal RF studies, showed broad transitions, suggesting that this may be a fundamental feature of the new superconductors. A superior batch of 4 crystals grown by ATT, Bell Labs showed a very sharp transition at 6 GHz as shown in Fig. 3 and compared with the result of the best bulk ceramic material studied at the same time. Below 80 K, the crystals do not significantly influence the Q of the Nb host cavity (2×10^6); the measurement sensitivity is limited by the small area (20 mm²) and by host cavity and sapphire holder losses. A separate measurement using a Nb holder for the crystals provided an upper bound of 15 $\mu\Omega$ for crystals at 3.5 K. Near T_c, the surface resistance drops by almost 500 between T_c and 0.9 T_c, a steeper drop than for Nb.

At higher RF fields, bulk YBCO shows an increase in Rf losses by an order of magnitude by 10 Oe. On the other hand crystals grown at Cornell as well as ATT showed less than a factor of 2 increase in losses up to fields as high as 70 Oe. Measurements at Argonne [1] (77 K, ~200 MHz) showed that although the RF resistance for bulk ceramic rods increased from .03% to 2% of the normal conducting value, no breakdown of the superconducting state was obtained up to 320 Oe, the maximum field that could be reached with the available power. With ATT crystals, it was possible to reach 90 Oe at 20 K.

Influence of Processing

With crystals, RF currents are confined to flow in the ab plane which is known to have superior superconducting properties (see next section). An epitaxial film grown with the c-axis in the plane of RF current flow showed a factor of 20 higher RF losses than with C-axis normal to the plane[8]. Crystals also eliminate the influence of grain boundaries. In these respects, crystal samples are closer to ideal YBCO. Results with crystals suggest that we can expect to see improvements in the RF resistance, as well as the high field behavior as material quality improves. Progress here is already forthcoming.

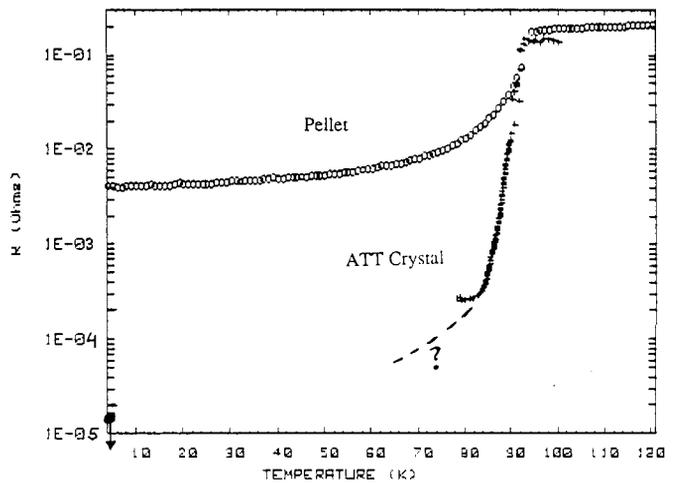


Fig. 3. The best 6 GHz RF transition on high quality single crystals from AT&T compared to the data taken ~ one year ago on a bulk ceramic pellet prepared at the U. of Wuppertal. In separate runs, two individual pieces showed R_s < 0.2 and R_s < 0.3 mΩ at 77 K. The rounding of the curve below 80 K is partly due to losses in the host cavity and crystal holder. A separate upper bound measurement of the 4 K resistance of four crystals is shown at 15 $\mu\Omega$ at the bottom left.

It has now become possible to produce sharp RF transitions in bulk ceramic material by introducing regrinding steps during calcination. Studies at the U. of Wuppertal[11] have found it possible to systematically lower the surface resistance of bulk ceramics, both at 3 and 21 GHz, by increasing the sintering time. X-ray fluorescence studies at Los Alamos[4] show that poor RF performance correlates with large fluorescence, emanating from microscopic regions of insulating phases. The beneficial effects of regrinding and increased sintering time were reproduced at Los Alamos and shown concurrently to reduce fluorescence[4].

Whether the best crystals at present represent intrinsic limits is still an open question. Most crystals still have a dense (0.1 μ m periodicity) structure of twin boundaries (see next section). Occasionally large (200 μ m x 200 μ m) twin free regions in crystals have been observed[12], showing that even twinning may some day be overcome. High quality crystal growing techniques are also in an evolutionary stage. Factors such as the type of flux used and residual flux, crucible contamination, crystal extraction technique, annealing time are all known to play a role.

Influence of Intrinsic Properties

In many respects, the intrinsic properties of the new HTS are very different from the familiar superconductors. Ultimately, these features could have a large bearing on RF properties, placing specific demands on fabrication processes to yield useful behavior for applicability to high performance RF cavities. The fundamental characteristics that may be a cause for concern are: short coherence length, anisotropy, and high sensitivity to detailed oxygen stoichiometry.

Coherence Length

The spatial extent of the superconducting wavefunction has a characteristic dimension, called the coherence length, ξ_0 . In the new superconductors, the coherence length is of order 10 Å. ξ_0 can be determined experimentally from H_{c2}(0) and the Ginzburg-Landau theory. Most measurements give $\xi_0 \sim 4$ Å along c-direction and 30 Å within the ab plane[13].

The small coherence lengths tell us that the transport properties will be extremely sensitive to minute defects, such as a grain boundary (gb) and its associated imperfections. Decoupling of superconducting grains will occur because the coherence lengths approach the spatial scale of the gb thickness, forming only weak links between individual grains. It is well known that in ordinary

superconductors, grain boundaries do not produce weak links. Any depression of the order parameter is averaged out over $\xi_0 \gg$ gb thickness. For a single phase, isotropic superconductor with a large coherence length, grain boundaries have little effect on the critical parameters.

For HTS, even at a "clean" grain boundary, the disorder that exists from breaking up of the unit cell could exceed the coherence length, especially in the case of the c-axis. Introduction of a second phase at a gb either as a discrete phase or as a gb segregation layer will serve to intensify the gb decoupling. Impurity atoms are often segregated to gb regions in many compounds. XPS studies on carefully prepared[14], nominally single phase YBCO material show O bound in BaCO₃ as areas of gbs exposed is increased. This occurs independently of whether BaO or BaCO₃ is used as starting material. On the other hand, high resolution e microscopy studies at LBL [15] do not show grain boundary contaminants, but there is evident an interruption of the 1-2-3 structure, with the intercalation of an extra atomic CuO layer. Such an interruption may alone be responsible for the weak links.

From these considerations, we can expect randomly oriented polycrystalline 123 to improve as processing techniques progress to the point when the spatial extent of the defects is decreased to a value close to the coherence length. For further improvements, texturing (ie orienting all the grains so that RF currents can flow only within the ab plane) may become essential, since the relevant coherence length will increase to ~ 30 A, and be more forgiving. On the other hand, texturing without removal of imperfections on a 30 A scale is unlikely to be helpful.

Anisotropy

There is a large anisotropy[13] of the magnetic and electrical properties between the c-axis and the ab planes, with superior behavior when current flow is in the ab plane. We have already mentioned the anisotropy of the coherence lengths. The lower critical field, Hc1, at which flux first penetrates is a factor of 10 lower when an external field is parallel to the ab plane than if the field is along the c-axis. Similarly Hc2, the critical field at which the flux penetration is complete, is 7 times higher for H parallel to the ab plane than H along the c-axis.

Both the larger coherence length and the better superconducting properties in the ab plane strongly indicate that for superior RF performance, it will be necessary to orient the grains so that the c-axis is everywhere normal to RF surface. It is already demonstrated feasible to deposit oriented thin films (μm) of YBCO on several dielectric substrates. These films show zero field dc critical current densities 3 - 4 orders of magnitude higher than polycrystalline material. Encouraging RF results are also forthcoming. The Stanford/UCLA group[8] has observed a factor of 100 decrease in 4K, RF losses at 100 GHz between oriented films and bulk ceramic.

Oxygenation

The superconducting behavior of deoxygenated Y₁Ba₂Cu₃O_{7- δ} materials as a function of δ show some interesting features. As oxygen stoichiometry is decreased from the ideal 7.0 to 6.7, [16] the Tc decreases smoothly from 91 to 77 K, and the normal state resistivity increases. Current results suggest that conduction in the n-state is due to 1-dimensional CuO chains. At O content below 7.0, chains are preferentially broken and conduction in the n-state decreases. In parallel, the coupling between low dimensional sub-structures is broken, degrading superconductivity which will still persists.

Estimate of Benefits from HTS for Future Accelerators

The maximum possible accelerating field achievable in a superconducting cavity is, in principle, limited by the so-called superheating field, Hsh. Hsh is related to and not usually too different from the thermodynamic critical field, Hc. For Nb, Hc is 2000 Oe, corresponding to an RF critical field limit of ~ 50 MeV/m for accelerating structures in use today. In view of the many theoretical uncertainties at this stage, it is not clear whether the same considerations apply to HTS. We assume that Hsh \sim Hc is still the relevant RF critical field to judge the impact.

Measurements of Hc₁ and Hc₂ on YBCO crystals show that Hc for HTS is likely to be as high as 27,000 Oe[13], ie more than 10 times higher than for Nb. Similar studies on Bi-compound (2212 phase, Tc = 84 K) show Hc = 10,000 Oe[17]. More direct measurements on early bulk ceramic YBCO samples near Tc (T/Tc=.97) already show a factor of 5 higher Hc than for Nb at the same temperature[18]. It is likely that the quality of early bulk ceramic samples are not as high as those of crystals. If we use the crystal results, the fundamental RF field limit is raised from 50 MeV/m for Nb to 400 MeV/m for YBCO. Realization of even a fraction of such fields would have a profound impact on the capital cost of a future large scale high energy physics facility, such as a TeV linear electron-positron collider.

One must bear in mind that the present day field levels in familiar Nb cavities are limited well below the 50 MeV/m fundamental limit by secondary mechanisms such as field emission when lower field limitations such as multipacting or thermal breakdown are avoided. Judging from the prevalence of field emission in cavities from various different materials, Cu, Nb, Nb₃Sn, Pb, it is clear that we will be unable to capitalise on the benefits of the enhanced fundamental capability of HTS materials, at least until the field emission problem is well under control.

The increased operating temperature of HTS could impact the capital cost, the operating cost as well as performance and reliability of accelerator cavities. Higher operating temperatures imply fewer cryogenic system problems, and the accompanying higher heat capacities would provide improved thermal stability. For a cost optimised machine, the lifetime (~ 10 year) operating cost will be roughly comparable to the capital construction. In assessing these impacts, we will assume that the operating temperature is at least a factor of 10 higher than in use today, based on the 90 K HTS transition temperatures compared with 9K for Nb. Operation at these temperatures will result in simplified refrigeration and cryostat, and is likely to reduce the cost of these components by a factor of 10 and 2 respectively. However these components constitute only a fraction of the total accelerator cost.

Bearing in mind the extreme unlikelihood that HTS will progress rapidly enough to impact a machine such as CEBAF, we use this example to estimate the net impact for a similar scale machine. For CEBAF[19], the refrigerator is estimated to constitute 15% and the cryostat 10% of the total machine cost (Linac, Recirculation Arcs, Beam Switch Yard, and Injector), so that the net likely benefit is to reduce the total machine cost by 15 - 20 %. The AC power demand to operate a 2 K refrigerator constitutes about 1/3 of the total power to operate the machine, the other components being the RF power source, the DC magnets in the recirculating arcs and the machine utilities. Thus the benefit from 20 K operation is likely to approach a 30% reduction in overall AC power consumption to run the machine.

As another example, we estimate the quantitative benefits for a future large scale machine, such as a fully superconducting TeV linear collider using the rough cost breakdown provided by refs. [20]. The capital cost is dominated by the structure, of which the cryostat costs are likely to constitute at most 30%. A factor of 2 reduction made possible by higher operating temperatures would therefore impact the overall cost of such a facility by 15%. Because of the 1% RF duty cycle, the refrigerator capital cost is reduced to below 5% the total capital cost, and the impact of high Tc here is minimal. For operating costs, the refrigerator associated AC power demand is again 1/3 the total power, so that a 30% reduction in operating cost can be anticipated from increased refrigerator efficiency at 10 times higher operating temperature.

Concluding Summary

Enthusiasm over the remarkable strides made in critical temperature are tempered with the difficulties in achieving useful properties. In the two years since the original discovery of HTS, remarkable progress has been made to understand the structure of the materials and to characterise some of their basic properties. In parallel, a vast array of experimental approaches in fabrication and processing have been tried. Y-, Bi-, and Tl- compounds are simultaneously under exploration. Between the multitude of approaches and the variety of compounds available, efforts are prone to suffer dilution, so progress could be retarded.

At 77 K, the best (crystal) surface resistance for YBCO is still a factor of 30 higher than for Nb at 4K. With the factor of 25 gain from Carnot efficiency between 77 and 4 K, even this level of resistance could prove useful, when achieved in a form and on a scale commensurate with cavity fabrication. A 500 MHz cavity with a Q over 10^8 at LN2 temperatures is indeed an attractive prospect. The current best 77 K value is only an upper bound. At 4K, the surface resistance of crystal YBCO becomes comparable to Nb (4K), so we can expect further improvement in the 77 K value. However, even for crystals, this residual resistance is a factor of 1000 higher than the typical residual resistance of Nb ($10^{-8} \Omega$).

The resistance of crystals was observed to be field independent up to 90 Oe at 20 K. In bulk material, the highest surface magnetic field reached while some YBCO remained superconducting is 320 Oe at 77 K. If realised together with low surface resistance, this could translate to a useful accelerating field (8 MeV/m), comparable to that achieved routinely today with full scale Nb structures, but is still far inferior to the best that has been achieved with Nb (1600 Oe).

Ignoring the time scale for achieving the necessary progress with HTS and with field emission, we emphasize that the potential benefits through higher field operation could be sizable for a future large scale accelerator. Through higher operating temperatures, we anticipate a significant, but non-overwhelming, improvement on the capital cost (15 to 30 %) and a similar (30%) impact on the operating cost both for an application that is adequately fulfilled by entrenched technology as well as for a future application.

Much work is in progress to produce potentially useful forms of HTS, such as bulk sintered ceramics or thick films on high thermal conductivity silver substrates. RF properties of these forms are still inferior to the best values quoted above. In particular, the surface resistance increases rapidly even at RF surface magnetic fields as low as 10 Oe. Some of the factors that could influence the poor RF quality of these HTS are just beginning to be understood. It is not yet clear whether the inferior RF properties observed so far are entirely process-dependent or whether they stem from some fundamental aspects peculiar to the new superconductors, such as the short coherence length, anisotropy and oxygen sensitivity. New experimental approaches to improve RF performance must address some of the problems understood, especially those that stem from the intrinsic nature of the new superconductors.

The technological effort necessary to overcome these difficulties may be substantial and incommensurate with the time scale for the next generation of accelerators. The example of Nb_3Sn may be a relevant parallel. Nb_3Sn with a Tc and Hc essentially twice that of Nb has been long known. Its potential advantages due to intrinsic properties have not been realised, presumably due to difficulties of forming the material with the required uniformity in surface stoichiometry. As discussed earlier, high performance RF is not very forgiving of imperfections. Performance of Nb_3Sn cavities significantly lags that of pure Nb, theoretically an inferior material.

Immediate impact on accelerators is not seen. Nb retains its superiority for applications in progress. Improvements in the performance of Nb cavities are also likely to yield further payoffs in the future. Accelerator cavities are not limited by the fundamental properties of Nb such as Hc or R_{BCS} , but by problems such as field emission. Progress in this area and with Nb cavities is an essential condition for realising the benefits of the higher fundamental fields that could become available as the current problems are solved with HTS. The fact that for pulsed fields surface RF fields of 400 MV/m have been achieved in copper gives hope that extensive development might overcome the field emission barrier.

It remains to be seen whether the new materials will rise to the challenge, but certainly exploration of the possibilities continue to be important.

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References

- [1] C. L. Bohn, J. R. Delaven, D. I. Dos Santos, M. T. Lanagan, and K. W. Shepard, Proc. of the 1988 Applied Superconductivity Conference, San Francisco, to appear in IEEE Trans. Magnetics (1989).
- [2] D. L. Rubin, K. Green, J. Gruschus, J. Kirchgessner, D. Moffat, H. Padamsee, J. Sears, Q. S. Shu, L. Schneemeyer, J. V. Wasczak, To Appear in Phys. Rev. B, Rapid Communications.
- [3] A. Fathy, D. Kalokitis, E. Belohoubek, To appear in Microwave Journal, October 1988 Issue.
- [4] W. Cooke, Los Alamos, National Lab., priv. comm.)
- [5] M.S. DiIorio, A. C. Anderson, B-Y. Tsaur, preprint R. Withers, priv. comm.
- [6] H. S. Newman, A. K. Singh, K. Sadananda, M. A. Imam, NRL, preprint, submitted to Appl. Phys. Lett
- [7] W. L. Kennedy and S. Sridhar, submitted to Solid State Comm. (1988)
- [8] T. L. Hylton, M. R. Beasley, A. Kapitulnik, J. P. Carini, L. Drabeck, G. Gruner, Proc. of the 1988 Applied Superconductivity Conference, San Francisco, to appear in IEEE Trans. Magnetics (1989).
- [9] B. R. McAvoy, G. R. Wagner, J. D. Adam, J. Talvacchio, M. Driscoll, Proc. of the 1988 Applied Superconductivity Conference, San Francisco, to appear in IEEE Trans. Magnetics (1989).
- [10] J. S. Martens, G.K. G. Hohenwarter, D. P. McGinnis, J. B. Beyer, D. S. Ginley, Proc. of the 1988 Applied Superconductivity Conference, San Francisco, to appear in IEEE Trans. Magnetics (1989).
- [11] G. Mueller, D. J. Brauer, R. Eujen, N. Klein, H. Piel and L. Ponto, Proc. of the 1988 Applied Superconductivity Conference, San Francisco, to appear in IEEE Trans. Magnetics (1989).
- [12] N. P. Ong, Z. Z. Wang, S. Hagen, T. W. Jing, J. claybold, and J. Hovrath, Physica C 153-155, 1072 (1988)
- [13] T. K. Worthington, W. J. Gallagher, and T. R. Dinger, Phys. Rev. Lett. 59, 1160 (1987).
- [14] A. Santoro, S. Miraglia, F. Beech, S. A. Sunshine, D. W. Murphy, L. F. Schneemeyer, and J. V. Wasczak, Mat. Res. Bull. 22, 1007 (1987).
- [15] H. W. Zandbergen, R. Gronsky, G. Thomas, Physica C 153-155, 1002 (1988).
- [16] S. I. Park, C. C. Tsuei, K. N. Tu, Phys. Rev. B (to be published).
- [17] B. Batlogg, T. T. M. Palstra, L. F. Schneemeyer, R. B. van Dover, and R. J. Cava, Physica C 153-155, 1062 (1988).
- [18] D. K. Finnemore et. al., Novel Superconductivity, ed. S. A. Wolfe & V. Kresin (Plenum) 1987.
- [19] CEBAF Design Report.
- [20] R. Sundelin, Proc. of the 1987 Particle Accelerator Conf., Washington DC., IEEE Cat. No. 87CH2387-9, p. 68.